PNL-4432-6
(FY 83 Fourth Quarter)

Interim Report

Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites

QUARTERLY PROGRESS REPORT
July-September, 1983

L. J. Kirby
W. H. Rickard
A. P. Toste

November 1983

Prepared for the
U.S. Nuclear Regulatory Commission
under Contract DE-AC06-76RLO-1830

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
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NRC FIN B2291

Pacific Northwest Laboratory
Richland, Washington 99352

PREFACE

This is the sixth quarterly report for the project "Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites" (NRC FIN B2291) under a reporting schedule initiated in mid-1982. The next report is scheduled in January, 1984.

The project task organization has undergone several revisions since this reporting schedule was established. To assist the reader to follow the progress of the research, as each new report is prepared we are summarizing previously reported research under a section titled "Summary of Prior Efforts."

Detailed reporting of research findings will continue to be published in quarterly, topical and annual reports and in the open literature.

EXECUTIVE SUMMARY

The primary purpose of this project is to develop an understanding of chemical processes that significantly influence the migration of radionuclides at commercial low-level waste burial sites. Chemical measurements of waste trench leachate and identification of chemical changes in leachate during migration will provide a basis for geochemical waste transport models. This project will produce for the U.S. Nuclear Regulatory Commission (NRC) information to support guidance for implementation of 10 CFR 61, particularly in the development of criteria for low level waste disposal site selection, management, permanent closure and monitoring. This project will also produce information needed by the Commonwealth of Kentucky as they finalize plans to stabilize, close and monitor the Maxey Flats site.

- Chemical Forms: Inorganic and Organic Radionuclide Species

  Comprehensive organic analyses performed on Maxey Flats waste trench leachate, water from the experimental trench and water from the inert atmosphere wells have determined a large number of hydrophilic and hydrophobic organic compounds in these waters. Analysis of the field-separated solutions from chromatographically fractionated water samples has revealed a complex pattern of association between radionuclides and organic compounds in these waters. Ethylenediaminetetraacetic acid (EDTA) is the major organic compound in water from waste trench leachates and in water from inert atmosphere well W2NA and evidence points to the migration of plutonium as an EDTA complex. Polar organic compounds (e.g. palmitic and stearic acids) may influence the migration of ^{90}Sr and ^{137}Cs.

  Analysis of waste trench leachate samples from waste trench sumps 27, 19W, 35, 7-3, 33L-4, 33L-8 and 23M at Maxey Flats in 1979 and 1981 revealed a complex variety of hydrophilic organic compounds. The chelating agent EDTA is present in all of the waste trenches, generally at parts-per-million (ppm) levels. The wide concentration range (0.078 to 12.4 ppm) for EDTA in waste trench leachates and even within the same waste trench emphasizes the high variability of leachate

chemistry from trench to trench, within the individual trenches and with time. EDTA-like species of lower molecular weights are present due to microbial diagenesis or radiolytic decomposition. N-hydroxyethylethylenediaminetriacetic acid (HEDTA) is also a major complexing agent in waste trenches 19S, 23 and 27.

Analysis of the hydrophobic organic compounds that were isolated to facilitate the chromatographic analysis of hydrophilic organic species has also permitted us to obtain information on toxic organic compounds that are present in these waters. Most of the hydrophobic organic compounds are present at parts-per-billion levels and only three of these compounds - pentafluorobenzoic acid (a non-radioactive tracer added to evaluate trench caps on the experimental trench), 2 (3H)-benzothiazolone (present in waste trench 27 leachate) and a group of alkylphenoxy oligomers (present in inert atmosphere well W2NA) - are present at parts-per-million levels. Two barbiturates, barbital and pentobarbital were also identified in water samples from waste trench 27 and inert atmosphere well W2NA.

- Subsurface Migration and Infiltration Studies

During the reporting period, we have continued sampling the porous cups, experimental trench sumps and inert atmosphere wells on a regular monthly basis, weather permitting. We have discontinued sampling the E-series wells around the Maxey Flats site; none of the samples collected contained significant radionuclide concentrations other than tritium. We also performed well-logging of inert atmosphere wells, experimental trench sumps, and three four-inch diameter E-series wells, confirming that waste radionuclide migration along the sandstone marker bed occurred only within the restricted area. These measurements verify that subsurface migration of radionuclides has occurred over limited distances within the Maxey Flats restricted area, but with the possible exception of tritium, subsurface migration to points outside the restricted area has not been a significant source of contamination by radionuclides of the environs adjacent to the Maxey Flats site.

Ecological Monitoring at Commercial Shallow Land Burial Sites

Ecological field sampling performed at Maxey Flats has evaluated a broad spectrum of radionuclides in leaves, forest floor litter and surface soil but only ^{3}H and ^{60}Co appear to be derived from the disposal site. Even these radionuclides are at only slightly elevated levels - well below the MPC's for drinking water. These surveys indicate that leaf fall can be a cost-effective, efficient way to estimate the annual contribution of radionuclides to the forest floor of oak-hickory forests. Tree sap and water from transpiring leaves have also been collected and analyzed for tritium, and indicate that tritium uptake by deeprooted trees has potential to serve as a biomonitor of subsurface movement of water from burial sites. These surveys have indicated that biomonitors may be useful as indicators of radionuclide migration from shallow land burial sites.

During the reporting period intensive tree leaf sampling was conducted outside the restricted area at Maxey Flats to determine whether tritium concentrations in individual trees changed as the growing season progressed and to evaluate the extent to which one tree containing higher-than-normal tritium concentrations was sampling its immediate vicinity. Tritium concentrations in leaf water tended to decline as the growing season progressed due to dilution of tritium in the groundwater the trees are utilizing. The data also pointed out that in the case of the tree containing higher-than-normal tritium concentrations the source of tritium is highly localized and this tree may pinpoint an ideal location to establish a monitoring well outside the restricted area.

Technical Program Coordination

The report PNL-4432-5, "Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites, Quarterly Progress Report, April-June, 1983 was published and distributed in July, 1983.

This project (B-2291) was reviewed at the Fifth Annual Participants' Information Meeting, U.S. Department of Energy Low-Level Waste Manage-

ment Program, Denver, Colorado, August 30-September 1, 1983. Two presentations were made, as follows:

L. J. Kirby and A. P. Toste, "Chemical Characterization, Migration and Fate of Radionuclides at Commercial Shallow Land Burial Sites".

W. H. Rickard and L. J. Kirby, "Radionuclides in a Deciduous Forest Adjacent to a Commercial Shallow Land Burial Site: Implications for Monitoring to Detect Radionuclide Migration".

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INTRODUCTION

One of the most important factors to be considered during the long-term isolation of radioactive wastes at commercial low level shallow land burial sites is the subsurface migration of radionuclides, which depends, among other things, on the chemical forms of the radionuclides. The oxidation state, ionic form, and the formation of complexes or chelates with organic ligands can markedly influence radionuclide migration. Therefore, determination of the precise chemical species of radionuclides that have migrated is essential for our accurate evaluation of the long-term performance of shallow land burial sites. It is also essential to identify the organic and inorganic compounds and ionic species which are present in buried waste or can be generated in situ by chemical reactions and subsequently participate in the formation of the chemical species that may be shown to migrate.

PNL has assisted in coordinating the NRC research program at Maxey Flats since 1979 to establish and carry out a unified and comprehensive study of the problems specific to that site and to obtain generic information that may be useful in setting criteria for decommissioning of existing commercial sites and for licensing of future commercial low level waste burial sites. The objectives of this broad, multidisciplinary, multilaboratory program include determining:

- Radiochemical and chemical composition of leachates in waste burial trenches;
- Subsurface migration rates of radionuclides and the chemical, physical, biological, and hydrogeological factors which affect this migration;
- Areal distribution of radionuclides at commercial shallow land burial sites and the factors responsible for this distribution;
- Concentrations of radionuclides in vegetation both on and off site and the uptake of radionuclides by representative agricultural crops;
- Atmospheric pathways for radionuclide transport and the mechanisms involved; and
- Engineering practices which influence the seepage of surface waters into the burial trenches.

OBJECTIVES

The primary purpose of this project is to develop an understanding of the chemical processes that influence radionuclide migration at commercial low-level waste burial sites. Chemical measurements of waste trench leachate and identification of chemical changes in chemical forms that occur during migration are being made and will provide a basis for geochemical waste transport models. This project will produce for NRC information to support guidance for implementation of 10 CFR 61, particularly in the development of criteria for commercial low level waste (LLW) disposal site selection, management, permanent closure and monitoring.

Specific objectives of this project with respect to measurements to be made at existing sites are:

- Define the actual chemical species of the radionuclides which are migrating by subsurface routes;
- Identify the chemical characteristics of the trench leachate which retard or enhance radionuclide mobility;
- Determine the chemical changes in leachate properties which result from installation of infiltration barriers such as the flexible vinyl covers at Maxey Flats;
- Provide the chemical and radiochemical input parameters essential for hydrological/geochemical modeling of subsurface radionuclide transport;
- Determine the factors responsible for the areal distributions of surface radionuclides and develop improved procedures for monitoring;
- Develop and test post-closure monitoring protocols that can be used at Maxey Flats and test the applicability of these protocols to operational and post-operational monitoring at other commercial sites; and
- Assist in coordinating LLW research projects for NRC.

Field measurements at shallow land burial sites provide unique opportunities to identify in actual waste burial situations the chemical and geochemical

parameters which retard or enhance radionuclide mobility at LLW burial sites. The work at Maxey Flats also responds to the need for information on the areal distribution of radionuclides and the extent of subsurface transport of radionuclides at that site. This data base is needed by the Commonwealth of Kentucky as they finalize plans to stabilize, close and monitor the Maxey Flats site.

SUMMARY OF PRIOR EFFORTS

Under this research program, PNL assists in coordinating NRC-sponsored LLW research, which has included PNL studies on chemical species and areal distributions of radionuclides, work on trench water chemistry by BNL, plant uptake studies by the University of California-Los Angeles, engineering studies by the University of Arizona, unsaturated flow studies by LANL and UCB, and water and mass balance studies by the Commonwealth of Kentucky.

To conduct experiments for determining the subsurface transport of radionuclides from existing waste burial trenches at a commercial LLW site, two experimental areas were established at the Maxey Flats, Kentucky, LLW disposal facility. This required the cooperation of several national and state agencies, together with the NRC contractors, for successful implementation. These experimental areas included:

- An experimental slit trench installed during the last quarter of FY 1979 and first quarter of FY 1980; and
- Fourteen inert atmosphere sampling wells installed in undisturbed soil adjacent to the experimental slit trench during the last quarter of FY 1980. A comprehensive array of porous cups and soil moisture cells was also installed for the LANL and UCB programs.

The experimental areas have been described in NUREG/CR-1832, Section IV and NUREG/CR-2383, Section I and are illustrated in Figures 1 and 2 of this report. These experimental areas have supported research studies by PNL, LANL, UCB and BNL.

The two experimental areas have enabled samples to be taken for the study of chemical species of radionuclides, saturated and unsaturated flow, nonradio-

active tracer movement, trench cap engineering and drainage, plant uptake, and total water balance by the laboratories participating in research at Maxey Flats. These experiments have provided needed information on rates and mechanisms of subsurface transport of radionuclides from waste burial trenches by both saturated and unsaturated flow and promise to provide more detailed information on subsurface flow at the site.

Field measurements have been performed by PNL at the Beatty, Nevada, (NUREG/CR-1524), and Maxey Flats, Kentucky, (NUREG/CR-2383, Section II and NUREG/CR-1832, Section IV) burial sites to help determine the areal distribution of radionuclides at these sites and the factors responsible for this distribution. Field measurements were also performed adjacent to the Maxey Flats facility, and confirmed that contamination is largely contained within the restricted area at this burial site. The areal distribution of radionuclides at Maxey Flats has been influenced by site operation and maintenance activities, surface contamination and runoff, deposition from the evaporator plume, and the actions of burrowing animals or deep-rooted trees. Vegetational and surface contamination on site and near site are quite low; of the gamma-ray-emitting radionuclides measured, only ^{60}Co exceeds commonly observed fallout levels. The average concentrations in surface soil at Maxey Flats are comparable to concentrations resulting from normal fallout in other areas of high rainfall. Tritium concentrations in water near the site have been influenced by surface runoff and by condensation from the evaporator plume.

Following analysis of trench leachates sampled in October, 1978 and November, 1979, Cleveland and Rees[13] concluded that except for leachate from waste trench 27 most of the plutonium in trench water is in true solution, in the tetravalent state, and exists in complexes with strong organic ligands. PNL research conducted in 1979 and 1980 (NUREG/CR-1832, Section IV), has demonstrated that plutonium in water from the experimental trench and in waste trench 27 leachate was present in highly complexed, anionic, reduced (trivalent and tetravalent) species. Ethylenediaminetetraacetic acid (EDTA) is the major organic complexing component in waste trench 27 leachate, but other polar, water-soluble organic compounds are also present (NUREG/CR-2383, Section I). Plutonium and EDTA co-elute during steric exclusion chromatography of Maxey

Flats trench and well waters, and evidence points to the migration of plutonium as an EDTA complex. Polar organic compounds (e.g. palmitic and stearic acids) may influence the migration of ^{90}Sr and ^{137}Cs. These studies have demonstrated the need to determine the stability of the plutonium-EDTA complex under the conditions existing in waste trench water at Maxey Flats, the conditions under which complexes of plutonium, americium, cobalt and strontium can be decomposed and/or sorbed on the geologic media, and the need to separate and measure the concentrations of hydrophilic organic complexing agents that may affect the migration of radionuclides present in waste trench leachate.

Comprehensive organic analyses performed on Maxey Flats waste trench leachate, water from the experimental trench and water from the inert atmosphere wells have determined a large number of hydrophobic organic compounds in these waters(2, 3). Analysis of the hydrophobic organic compounds that were isolated to facilitate the chromatographic analysis of hydrophilic organic species has also permitted us to obtain information on toxic organic compounds that are present in these waters(2). Most of the hydrophobic organic compounds are present at parts-per-billion levels and only three of these compounds -pentafluorobenzoic acid (a non-radioactive tracer added to evaluate trench caps on the experimental trench), 2 (3H)-benzothiazolone (present in waste trench 27 leachate) and a group of alkylphenoxy oligomers (present in inert atmosphere well W2NA) - are present at parts-per-million levels. Two barbiturates, barbital and pentobarbital were also identified in water samples from waste trench 27 and inert atmosphere well W2NA.

Changes in tritium concentrations in water from the experimental trench and in inert tracers (e.g., sodium bromide) which were added to the trench (NUREG/CR-1832, pages IV-10 and IX-6), confirm that subsurface flow does occur rather rapidly within the limited area of these experimental facilities. While increases in water level have occurred in the experimental trench, the absence of tracers in the water indicates that these increases have occurred without movement through the different trench caps installed on the five sections of the experimental trench (NUREG/CR-2383, Section I).

Ecological field sampling performed at Maxey Flats (NUREG/CR-1832, Section V, and NUREG/CR-2383, Section III) has evaluated a broad spectrum of

radionuclides in leaves, forest floor litter and surface soil, but only ^{3}H and ^{60}Co appear to be derived from the disposal site. Even these radionuclides are at only slightly elevated levels and are comparable to ambient fallout levels. Collections of freshly fallen tree leaves indicate that leaf fall collection is a cost-effective, nondestructive sampling procedure that can be incorporated into ecological monitoring programs to indicate migration or lack of migration from shallow land disposal sites. Tree sap and water from transpiring leaves have been collected and analyzed for tritium, indicating that tritium uptake by deeprooted trees has potential to serve as a biomonitor of subsurface movement of water from burial sites. The field surveys have indicated that biomonitors may be useful as indicators of radionuclide migration from shallow land burial sites.

Carbon-14 analyses of wood from selected trees adjacent to the Maxey Flats restricted area were compared with analyses from tree specimens at a location 12 miles from the Maxey Flats Facility. These limited data suggested that the site may have influenced ^{14}C levels very slightly at a few locations near the perimeter fence. The source of ^{14}C in the wood specimens is from carbon dioxide that is assimilated from air in the photosynthesis process(1).

Remaining important areas of research include the fate of mobile (possibly complexed) radionuclide species after leaving the trench environment, the control of these species within the commercial site boundaries, remedial action that can be taken to halt this mobility, and whether complexing agents such as EDTA can be effectively destroyed in an aerobic environment.

Important observations from research conducted by PNL at Maxey Flats include:

- Identification of the magnitude of transport over short distances within the burial site by subsurface flow;
- Characterization of some of the important chemical species including plutonium-EDTA complexes which are responsible for subsurface migration;
- Identification of surface runoff from the site as a principal source for off-site radionuclide accumulation in vegetation;

- Determination of the off-site distribution of radionuclides at Beatty, Nevada, and Maxey Flats, Kentucky; and
- Establishment of the complexity and variability of waste trench leachates as source terms for the migration studies. (PNL research complements the comprehensive analyses performed on waste trench leachate by BNL. See, for example, NUREG/CR-2383, Section IV, and NUREG/CR-1832, Section III.)

These research observations and related observations from the research performed at Maxey Flats by BNL, LANL, UCB, the University of Arizona and the University of California-Los Angeles are described in NUREG/CR-2383[10] and NUREG/CR-1832[16]. Reports and summaries of PNL research are contained in the References section of this report.

SIGNIFICANT CURRENT RESEARCH RESULTS

This is the sixth quarterly report for the project "Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites" under the revised reporting schedule requested by NRC. The organization of the third, fourth and fifth reports has been changed slightly from that of the first two quarterly reports[4-5] to conform to the revised research proposal format. We will continue to report and summarize research results by subject and task.

Task A - Chemical Forms: Inorganic and Organic Radionuclide Species

The migration rates of radionuclides depend to a major degree on their chemical species. It is therefore extremely important that we determine chemical species in the waste burial trenches and in any water which has carried these radionuclides to our sampling points in the experimental trench and inert atmosphere sampling wells, or to springs some distances from the waste-filled trenches. In this task we are determining as nearly as practicable the precise chemical forms for those radionuclides which are of greatest concern from an environmental and health standpoint. We expect to report the predominant oxidation state of plutonium, and whether it is present as a complex, a chelate, or as a simple ion. Similar identification will be

made for other radionuclides, e.g. ^{241}Am, ^{60}Co, ^{137}Cs and ^{90}Sr. The precise radiochemical and chemical parameters obtained from these studies will provide important basic information on the chemical species that develop under various burial conditions. In these studies we will continue to supplement our laboratory work with on-site chemical separations employing tracers and organic ligand separations. The on-site separations minimize solution degradation and thereby duplicate as closely as possible actual field conditions. These studies will serve as a basis for modeling of the Maxey Flats site.

During the reporting quarter we have performed a further detailed analysis of the EDTA-like species that were measured in waste trench leachates and inert atmosphere well water at Maxey Flats(2, 3). We previously identified three EDTA-like species of molecular weights 219, 244 and 288 with structures similar to EDTA and related chelating agents (e.g. NTA, DTPA and HEDTA). We suggested that these EDTA-like species might be derived from the degradation of EDTA and assigned tentative structures to the EDTA-like species.

Following additional detailed examination of our data and further laboratory study, we conclude that the molecular weight 288 species is derived from the chelating agent HEDTA and not from EDTA as previously assumed. Based on gas chromatography (GC) and GC-mass spectrometry (GC-MS) anlayses of chelating agents and of the Maxey Flats groundwater samples, we propose that the molecular weight 288 species arises as a procedural side-product of the methylation reaction used in our groundwater analysis scheme. The proposed mechanism for its formation is outlined in Figure 3. Using appropriate GC response factors for methylated HEDTA we have requantified the molecular weight 288 species as HEDTA. See Table 1. HEDTA appears in leachate from four sampling sumps (19S, 19W, 23M and 27). The relative amount of HEDTA varies considerably, presumably as a result of differences in the wastes originally buried and/or differential degradation of the chelating agents. In waste trench 23M the HEDTA/EDTA ratio is quite high (4.0) whereas in waste trench 27 it is relatively low (0.07).

The detailed chemical speciation studies have demonstrated that EDTA is co-eluting with plutonium and ^{60}Co during steric exclusion chromatography of water from waste trenches 27 and 19S and inert atmosphere well W2NA. Strontium-90 and ^{137}Cs also appear to be associated with polar organic compounds,

notably palmitic (hexadecanoic) and stearic (octadecanoic) acids. Quantitation of EDTA and of radionuclides in the concentrated and fractionated samples has demonstrated that the soluble plutonium is associated with EDTA.

Some of the plutonium originally present in the water samples is removed from the water during concentration of the samples by co-precipitation with iron. This occurs due to exposure to traces of oxygen or because the solubility limits for iron have been exceeded in the concentrated samples. This observation suggests that uncomplexed plutonium will be fixed to the soil with the iron hydroxide precipitate when the anoxic groundwater is exposed to oxidizing conditions.

Task B - Subsurface Migration and Infiltration Studies

The experimental trench and wells study was undertaken to determine if subsurface migration of radionuclides had occurred at Maxey Flats, to measure the extent of any such migration, and to define the processes by which movement occurs. The work is continuing to utilize the experimental installations we established on the Maxey Flats site for defining migration rates and specific chemical species of long-lived fission products and the transuranium elements. It is also allowing the determination of the chemical species of migrating nonradioactive elements associated with waste and utilizes nonradioactive tracer movement to evaluate the effects of engineered trench caps and drainage systems on water infiltration rates.

During the reporting period, we have continued sampling the porous cups, experimental trench sumps and inert atmosphere wells (Figure 2) on a monthly frequency, weather and water levels permitting, and analysis of the samples for radionuclides and organic compounds is continuing. The water samples are analyzed for ^{3}H, gamma-ray emitting radionuclides (e.g., ^{60}Co, ^{137}Cs), alpha particle emitting radionuclides (e.g., $^{238,239,240}Pu$, ^{241}Am), hydrophilic and hydrophobic organic species, and nonradioactive elements that can be determined by neutron activation (e.g., Br). These measurements verify that within the Maxey Flats restricted area subsurface migration of radionuclides has occurred over limited distances (NUREG/CR-2383, page I-13). Except for tritium, migration to points outside the restricted area has not been a significant source of contamination by radionuclides.

During the reporting quarter we performed well-logging of our inert atmosphere wells and experimental trench sumps (Figure 2). We also performed well-logging of wells 13E, 14E and 3E (Figure 4). Only one location, well 14E in the central part of the restricted area, showed evidence that waste-derived radionuclides such as ^{60}Co and ^{90}Sr had migrated. These radionuclides were detected at the surface of the sandstone marker bed and the data are now being quantitated. These measurements again confirm that, except for tritium, migration of radionuclides has been confined mainly within the restricted area at Maxey Flats.

Task C - Specific Radionuclide Mapping at Maxey Flats and Other Commercial Shallow Land Burial Sites

The Maxey Flats burial site has been contaminated by several sources. These include the overflow of water from the burial trenches as rainwater infiltrated and filled them, spillage during pumping of excess water from the trenches, fallout from entrained radionuclides in the evaporator plume, and surface runoff of radioactive solutions from the site. To obtain a better understanding of the actual sources of radioactivity in surface soils and vegetation on the burial sites and adjacent areas, a detailed study of the levels of radioactivity has been underway at Maxey Flats and related studies should also be useful at other commercial shallow land burial sites.

In situ gamma ray surveys conducted at the Beatty, Nevada (NUREG/CR-1524), and Maxey Flats, Kentucky (NUREG/CR-2383, Section II and NUREG/CR-1832, Section IV) shallow land burial sites confirmed that most of the waste radionuclides have been retained within the boundaries of those sites. The in situ measurements were correlated with and augmented by analysis of samples taken from each of the counting locations, since only the more energetic gamma ray emitting radionuclides are measured by the in situ procedures.

Slightly elevated tritium concentrations in water samples from springs and seeps outside the Maxey Flats restricted area suggested that other radionuclides might also be slightly elevated at these locations. However, all gamma-ray measurements have indicated that ^{60}Co and ^{137}Cs are present at

ambient fallout levels. Strontium-90 and ^{238}, ^{239}, ^{240}Pu were also at ambient fallout levels, as determined from radiochemical separation and analysis of aliquots of soil and water. The measurements indicate that radionuclide migration to these springs and seeps is not occurring, except possibly for tritium.

These studies have demonstrated that surface contamination by radionuclides has been retained mainly within the restricted area at Maxey Flats. Surface sediments in areas adjacent to the site, primarily on the west side, contain ^{60}Co in concentrations greater than fallout. The origin of the ^{60}Co is from surface runoff into the adjacent woods. Measurements made to date indicate that offsite contamination has not resulted from subsurface transport, except possibly for tritium. The relative contributions of tritium from air (evaporator output), surface and subsurface pathways are being further evaluated during the second six-month curtailment of evaporator use by the Commonwealth of Kentucky.

Task D - Ecological Monitoring at Commercial Shallow Land Burial Sites

In this task we are developing efficient and statistically valid ecological field sampling procedures and methods for post-closure monitoring at shallow land burial sites. Kinds and amounts of radionuclides and trace elements in environmental samples are being measured with emphasis on biotic uptake, bioaccumulation, biotic transport, and ecological pathways in semi-wild ecosystems. We are emphasizing the sampling for pattern approach to provide three dimensional data that are amenable to rigid statistical analysis.

Forest sampling at Maxey Flats indicates the presence of a broad spectrum of naturally occurring and fallout-derived radionuclides in the litter layers, surface soil, and in freshly fallen autumn leaves(11). Radiochemical analyses of gamma-ray emitting radionuclides show very low levels of all radionuclides and only ^{60}Co was elevated above ambient fallout levels and is most likely of disposal site origin. Collections of freshly fallen tree leaves indicate that leaf fall collection is a cost-effective, nondestructive sampling procedure that can be incorporated into ecological monitoring programs to indicate migration or lack of migration from shallow land disposal sites.

Tree sap and water from transpiring leaves have also been collected and analyzed for tritium; the results indicate that tritium uptake by deep-rooted trees likewise has potential to serve as a biomonitor of subsurface movement of water from burial sites. Tritium is environmentally one of the most mobile of radionuclides and it can move as tritiated water, as vapor, or in surface or groundwater flows. Trees obtain water from their rooting substrate and move it from roots to stems to leaves, and release water to air as vapor. Water can be extracted from fresh tree leaves and analyzed for tritium. If tritium is present in the root zone, it appears in the extracted water. Because trees have deeply penetrating roots, as compared to crop plants, they can be used to monitor subsurface water flows. Leaf water data indicate that tritium in above ambient fallout levels is present within the edge of the forest at Maxey Flats(2, 3). Maple trees, Acer sacharum, have the unusual ability to move water through their trunk before new leaves emerge in the spring. If maple tree roots have access to tritiated water, tritium will appear in the sap stream. Experience at Maxey Flats indicates that sap samples for tritium analyses can be obtained by tapping tree trunks in a cost-effective and a biologically nondestructive fashion useful for post closure ecological monitoring programs.

Autumn leaf-fall samples were collected from the 0.5 m^2 rectangular wire baskets placed beneath the permanently tagged trees adjacent to the Maxey Flats restricted area during the first quarter, FY 1983. These samples have been dried and shipped to our PNL facilities at Richland, Washington, for analysis by gamma-ray spectrometry. We have determined the exogenous radionuclides (e.g., ^{60}Co, ^{137}Cs) and obtained information on endogeneous (e.g., ^{7}Be, ^{40}K, ^{226}Ra, ^{228}Th) radionuclides in some of the samples. Comparison of their radionuclide concentration with values reported from earlier sampling runs (NUREG/CR-2383, Section III, and NUREG/CR-1832, Section V) indicate again that buried waste radionuclides have not migrated into the forest adjacent to the Maxey Flats site. These analyses will be completed during the next several months.

Maple trees have now been tapped at more than 50 locations around the Maxey Flats disposal site and two trees have been tapped at a control location

in a similar forest type near Cave Run Lake, about 12 miles south of Maxey Flats. Tritium concentrations in tree sap ranged from below detection level to 290,000 pCi per liter of tree sap. The highest values occurred in two trees located along the western side of the disposal site and the lowest values were measured near Cave Run Lake, which represented fallout (background) tritium levels. Trees sampled in the vicinity of the disposal site generally had tritium values higher than fallout levels, with the highest concentrations being measured on the steep slope bordering the western side of the disposal site. In general, tritium values declined with increasing distance from the disposal site.

Two sampling trips were made early in the fourth quarter, FY 1983, to evaluate the feasibility of using extractable leaf water from tree species other than maple as an indicator of tritium migration. Tritium concentrations were determined in leaf water following an intensive sampling conducted outside the restricted area at Maxey Flats in July, 1983. Many of the same trees along the western side of the restricted area were resampled in August, 1983 to determine whether tritium concentrations in individual trees changed as the growing season progressed. The August sampling trip also permitted a more thorough evaluation of one area about 50 meters outside the fenced restricted area to be made.

Individually numbered trees located outside the restricted area were generally 8 to 20 meters from the fence, although one tree in the west drainage was as close as 3 meters and some locations along the north side were about 100 meters from the fence. The relative locations of the trees sampled are shown in Figure 5. In addition to the trees sampled around the fence, a transect was also made from the fence westerly into Drip Springs Hollow, with samples taken at 50-meter intervals, as summarized in Figure 6. Background samples were taken near Cave Run Lake, about 12 miles south of the Maxey Flats facility. Tritium concentrations determined after the July sampling trip are summarized in Table 2.

Tritium concentrations in leaf water tended to decline as the growing season progressed, as illustrated in Table 3. The decline may be due to the additional time between samplings, extending the time since evaporator shutdown.

Further dilution of the tritium contained in the groundwater that the trees were utilizing could have occurred during this time. The decline might also be an artifact of the unseasonably dry weather that prevailed at the Maxey Flats site during this sampling period.

One tree sampled along the transect into Drip Springs Hollow in July, 1983, contained much higher than normal concentrations of tritium. Tree number 055 contained the highest concentration of tritium and was resampled during the August, 1983 sampling trip. In July, tree 055 contained tritium at a concentration of 3.62×10^6 pCi/ℓ. In August this same tree contained tritium at a concentration of 1.25×10^7 pCi/ℓ. Tree number 097 in the vicinity of tree 055 contained tritium at a concentration of 2.69×10^7 pCi/ℓ. Other trees near this location 50 meters downslope from the fenced area contained substantially lower tritium concentrations, indicating that the source of tritium tapped by trees 055 and 097 is highly localized. These trees may pinpoint an ideal location to establish a monitoring well outside the restricted area.

Carbon-14 is present in the waste trenches at Maxey Flats and is one of the radionuclides of concern for ecological monitoring following site closure (10 CFR 61). Although ^{14}C has been widely used as a tracer in biological experiments, comparatively little is known about the root uptake of ^{14}C from soil or soil water. Carbon-14 analyses on selected samples of wood obtained from trees adjacent to the Maxey Flats restricted area were compared with analyses from trees located about 12 miles south of Maxey Flats facility, in the Daniel Boone National Forest. These limited data suggest that the site may have influenced ^{14}C levels very slightly in a few locations near the perimeter fence, where ^{14}C concentrations are slightly elevated above the "modern value," which is in the range of 8-8.5 pCi/gm. The source of ^{14}C in wood is from carbon dioxide assimilated from air in the photosynthesis process.

Task E - Technical Program Coordination for Low-Level Waste Research

The report PNL-4432-5, "Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites, Quarterly Progress Report, April-June, 1983" was published and distributed in July.

This project (B-2291) was reviewed at the Fifth Annual Participants' Information Meeting, U.S. Department of Energy Low-Level Waste Management Program, Denver, Colorado, August 30-September 1, 1983. Two presentations were made, as follows:

L. J. Kirby and A. P. Toste, "Chemical Characterization, Migration and Fate of Radionuclides at Commercial Shallow Land Burial Sites".

W. H. Rickard and L. J. Kirby, "Radionuclides in a Deciduous Forest Adjacent to a Commercial Shallow Land Burial Site: Implications for Monitoring to Detect Radionuclide Migration".

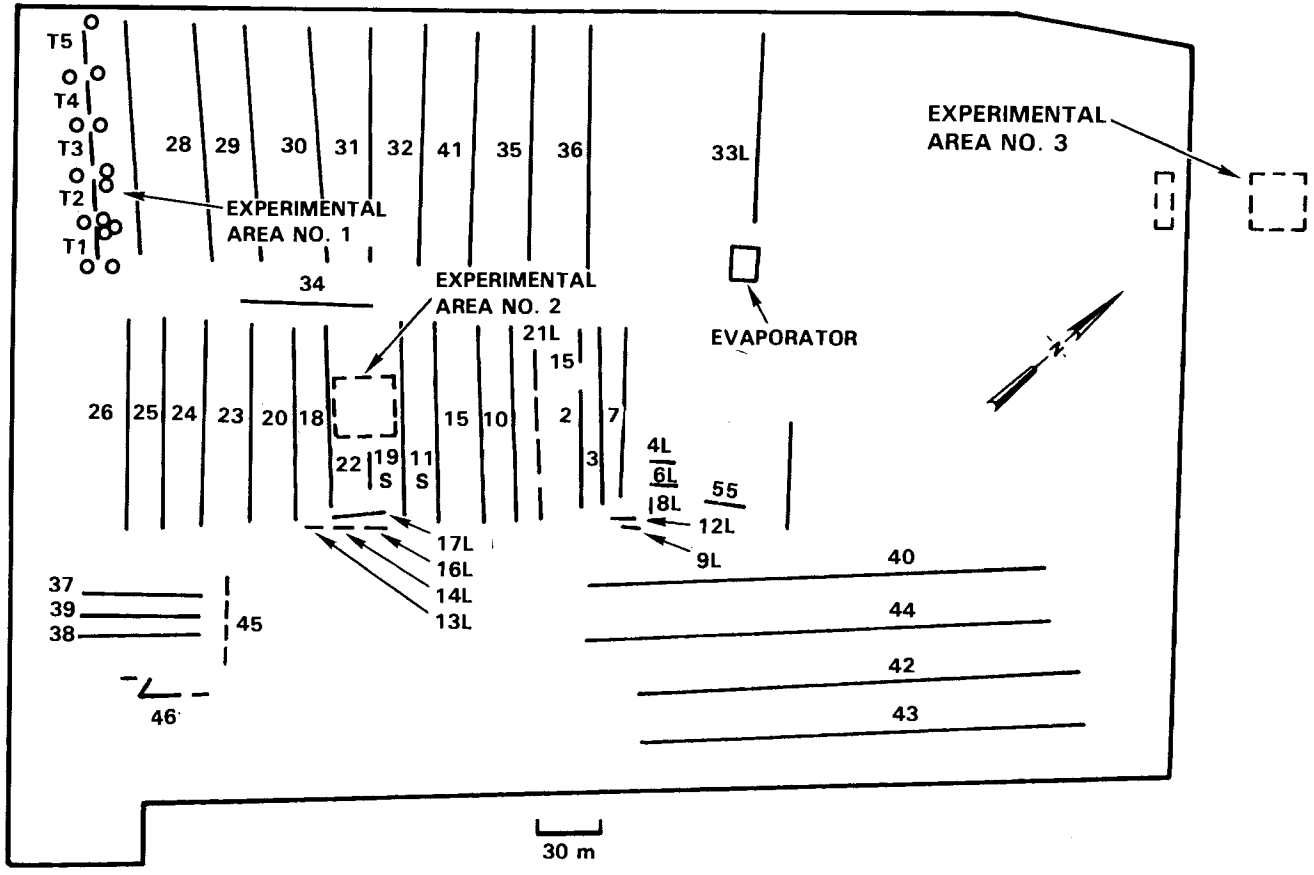

FIGURE 1. Maxey Flats waste trenches and experimental areas. The numbered lines show the approximate locations of the waste trenches. Experimental area number 1 includes the experimental trench (T1 through T5) and inert atmosphere wells (open circles). Experimental area number 2 includes the porous cup and tensiometer installations for studying water movement into trenches and water and radionuclide movement out of trenches. Experimental area number 3 includes the UCB lysimeter installations for water balance studies. These experimental areas support research studies by PNL, LANL, UCB and BNL.

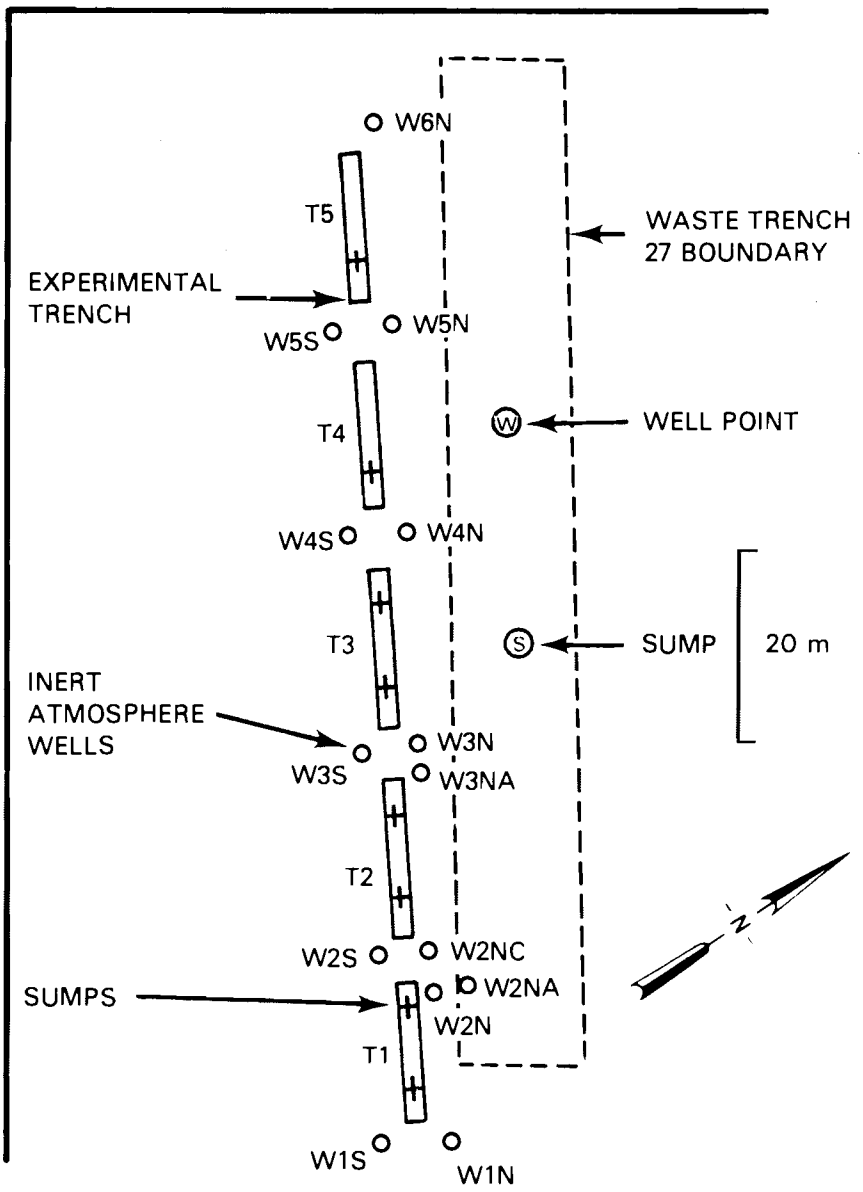

FIGURE 2. Maxey Flats Experimental Area No. 1, containing the experimental trench and wells near waste trench 27. These facilities allow concurrent sampling of the experimental trench, inert atmosphere wells and adjoining waste trench for PNL groundwater migration and chemical species studies. The experimental trench is also used by LANL. UCB, and BNL in research studies at the Maxey Flats Shallow Land Burial Site.

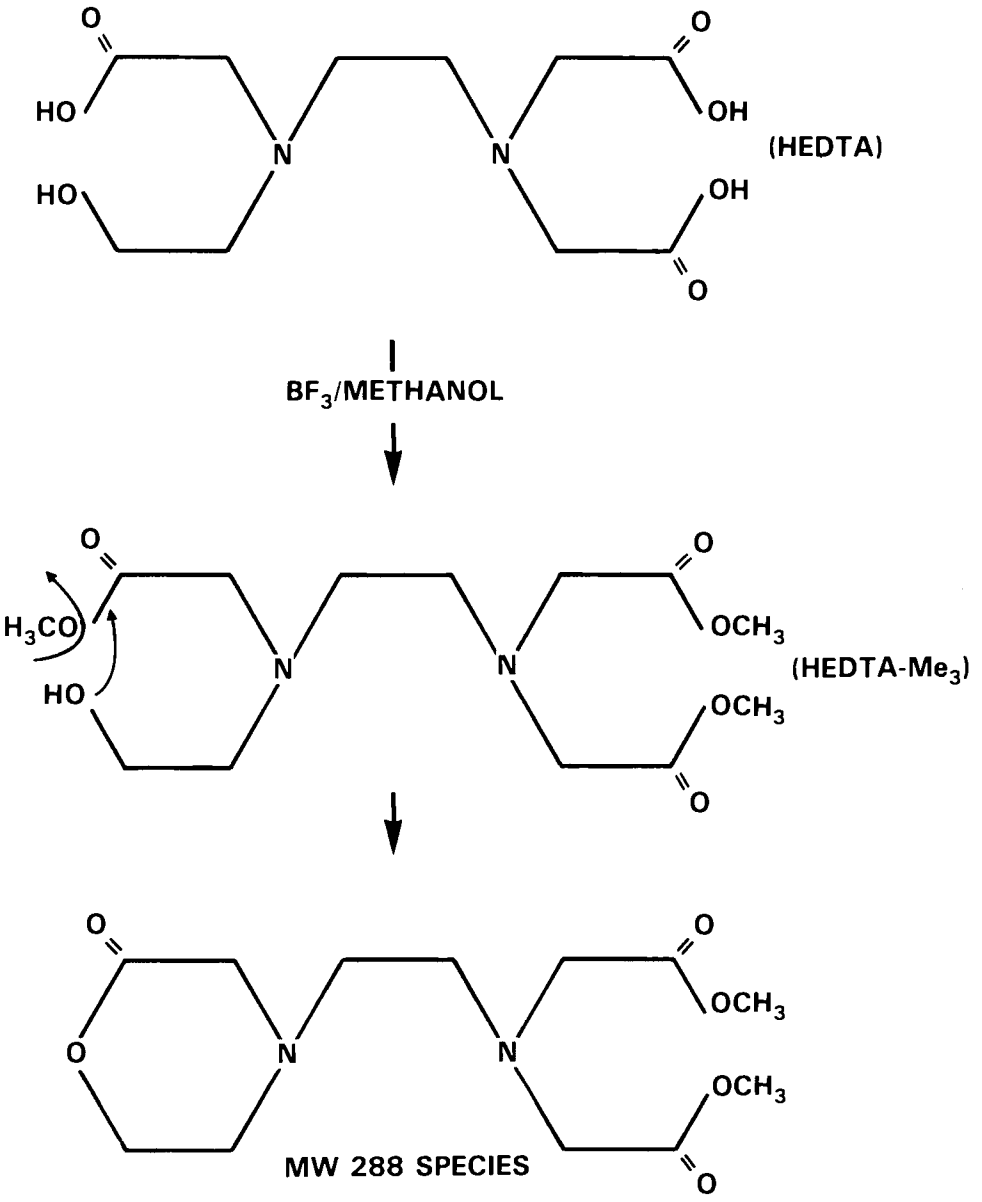

FIGURE 3. Formation of molecular weight 288 species from methylation of HEDTA. The MW 288 species probably arises as a procedural side product of the methylation reaction used in the groundwater analysis scheme. This particular species is probably not present as such in waste trench leachate.

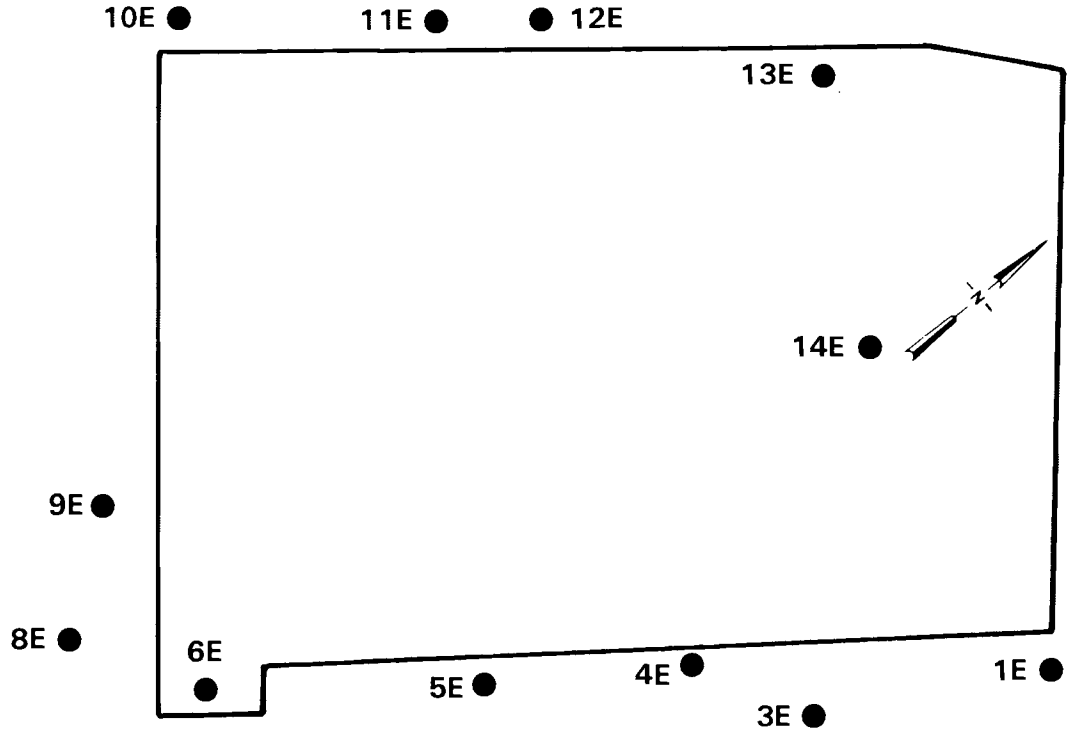

FIGURE 4. Relative locations of the E-series wells around the Maxey Flats restricted area. Wells 5E, 12E and 13E received large volume sampling to provide information on any radionuclides which migrate for significant distances and demonstrated that except for tritium any radionuclide migration has been confined mainly within the restricted area. Three four-inch diameter wells-13E, 14E and 3E - were logged and only 14E showed evidence of the presence of waste-derived radionuclides, e.g. ^{60}Co and ^{90}Sr. These radionuclides had migrated short distances within the restricted area along the sandstone marker bed.

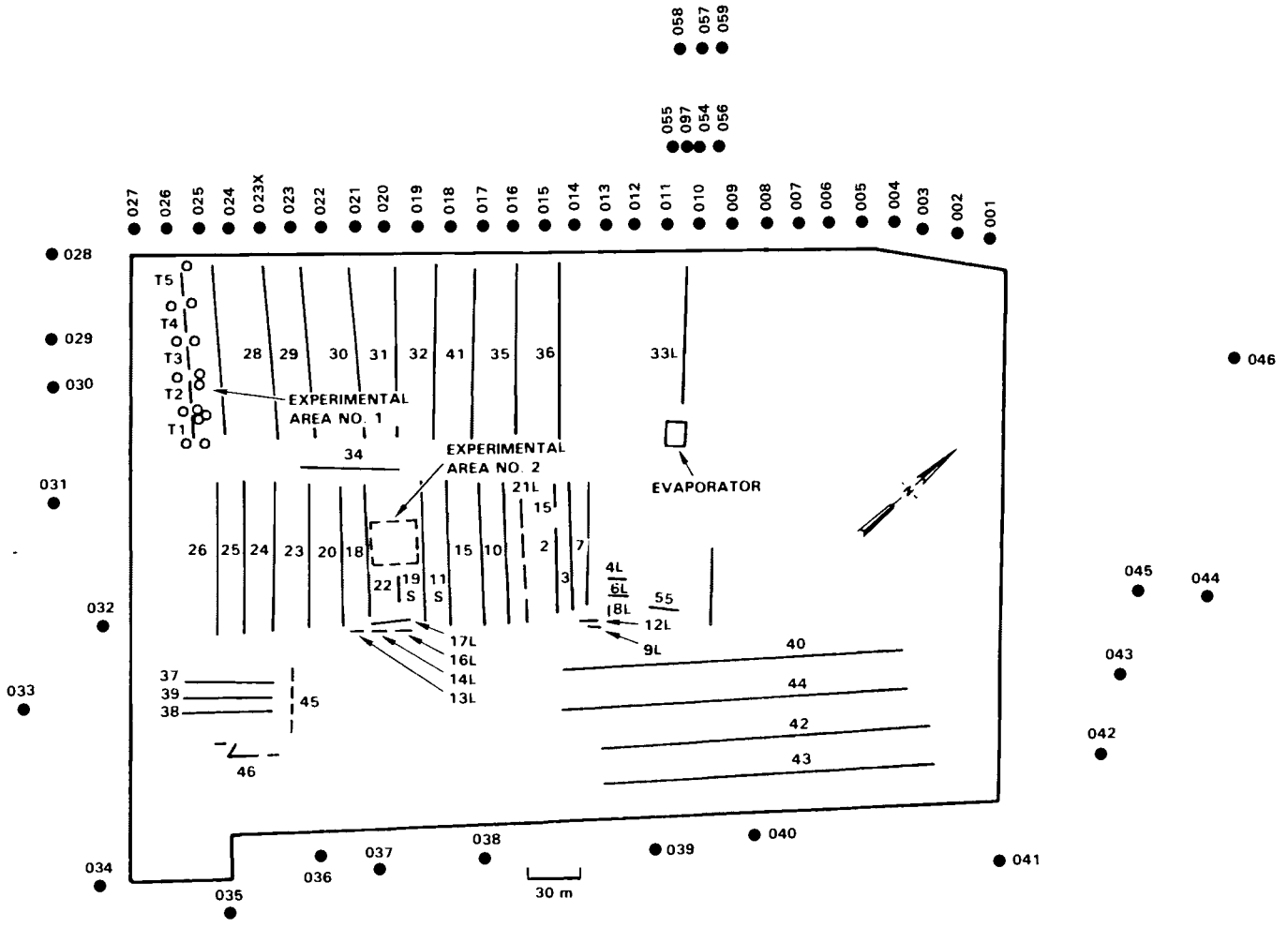

FIGURE 5. Relative locations of trees sampled outside the restricted area at Maxey Flats, July and August, 1983. These trees were generally located 8 to 20 meters outside the fence. Trees were also sampled at greater distances from the fence, and only two trees, numbers 055 and 097, contained higher-than-normal tritium concentrations.

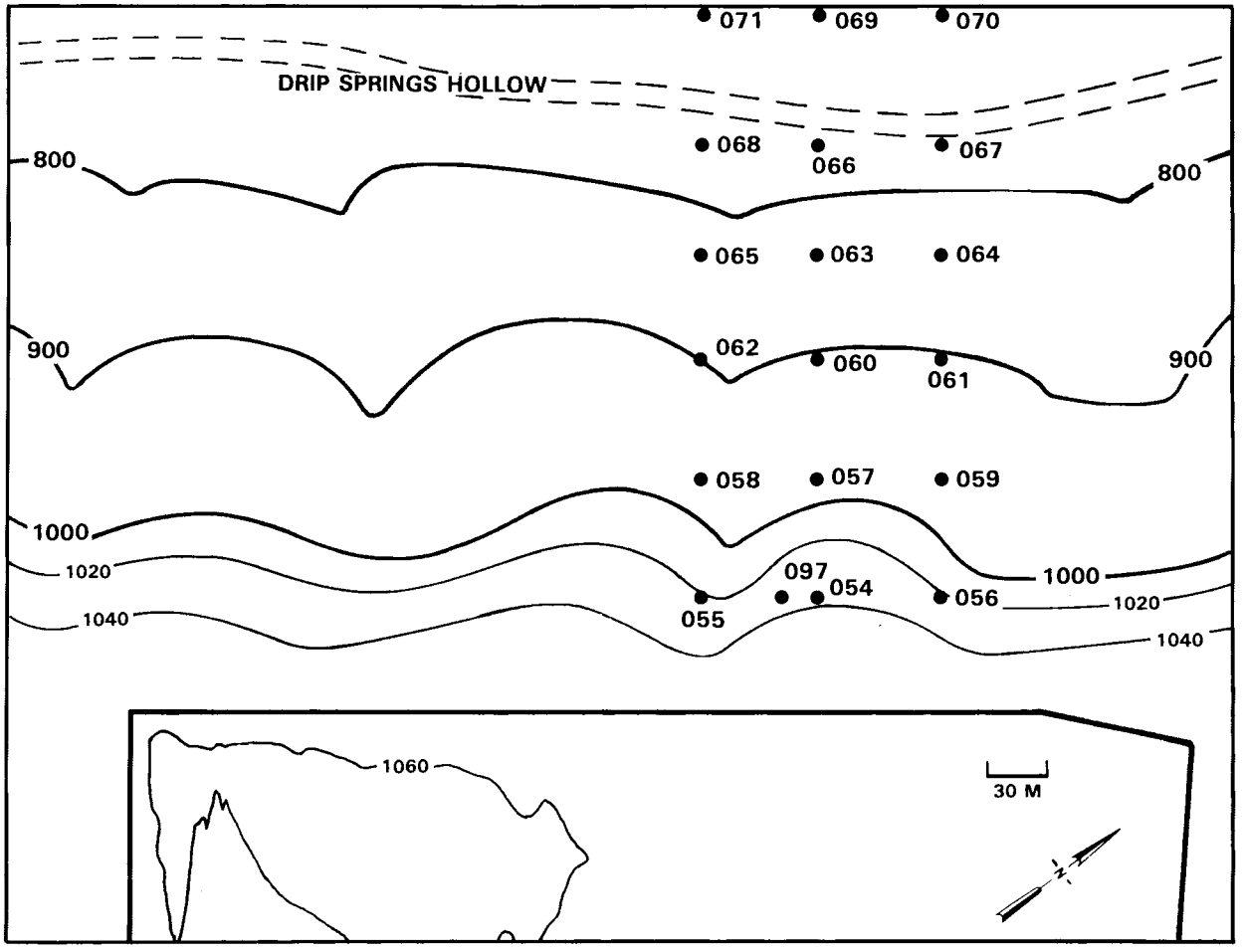

FIGURE 6. Relative locations of trees sampled in the west drain outside the restricted area at Maxey Flats, July and August, 1983. Trees 055 and 097 contained higher-than-normal tritium concentrations.

TABLE 1

Identification of HEDTA in Leachate from Waste Trenches at the Maxey Flats Shallow Land Burial Site

	HEDTA Concentration (ppb)			
	Waste Trench 19S (7-22-82)	Waste Trench 19W (10-81)	Waste Trench 23M (10-81)	Waste Trench 27 (7-27-82)
N-Hydroxyethylethylene-diaminetriacetic acid (HEDTA)[a]	888	8,350	19,511	285
Ethylenediaminetetraacetic acid (EDTA)[a]	4261	12,364	4895	3830
Chelator Fragment[a] (MW 244)	823	2,190	3884	758
Chelator Fragment[a] (MW 219)				trace

a) Methyl Ester (methylated with BF_3/Methanol)

TABLE 2

Tritium Concentrations in Leaf Water from Individual Trees Outside the Restricted Area at Maxey Flats, July, 1983 (See Figure 5 for Relative Tree Locations)

Tree No.	Species	^3H, pCi/ml	Tree No.	Species	^3H, pCi/ml
001	Oak	8.68	022	White Oak	28.1
002	Hickory	18.8	023	Hickory	8.01
003	White Oak	12.5	023X	White Oak	17.4
004	White Oak	18.2	024	White Oak	11.3
005	White Oak	15.8	025	Hickory	12.5
006	White Oak	22.5	026	Hickory	10.4
007	White Oak	321	027	Chestnut Oak	10.3
008	White Oak	21.5	028	Red Oak	4.08
009	Hickory	14.6	029	Cherry	5.23
010	White Oak	32.1	030	Hickory	2.24
011	White Oak	20.9	031	White Oak	7.23
012	White Oak	12.0	032	White Oak	4.54
013	Maple	17.3	033	Hickory	1.05
014	Black Locust	13.5	034	Chestnut Oak	6.18
015	Hickory	8.48	035	White Oak	4.32
016	White Oak	13.2	036	White Oak	6.37
017	Oak	10.3	037	Hickory	5.22
018	Red Oak	12.4	038	White Oak	38.4
019	Black Locust	14.7	039	Cherry	5.42
020	White Oak	56.9	040	Maple	10.9
021	Hickory	14.8	041	Oak	6.94

TABLE 2 (continued)

Tree No.	Species	^3H, pCi/ml	Tree No.	Species	^3H, pCi/ml
042	Red Oak	8.32	063	Maple	2.59
043	Oak	12.3	064	Maple	2.83
044	Maple	8.13	065	Maple	2.82
045	Box Elder	8.19	066	Maple	8.10
046	White Oak	7.15	067	Hickory	9.12
054	Hickory	27.1	068	Tulip Tree	4.58
055	Red Oak	3520	069	White Oak	3.88
056	Hickory	92.8	070	Maple	3.43
057	Oak	11.5	071	White Oak	4.70
058	Maple	3.93	075	Red Oak[a]	*
059	White Oak	11.9	076	Black Oak[a]	*
060	Chestnut Oak	6.06	077	White Oak[a]	*
061	Maple	4.23	078	Hickory[a]	*
062	Red Oak	7.25	079	White Oak[a]	*

*Background samples from Cave Run Lake are below detection level.

TABLE 3

Tritium Concentrations in Leaf Water from Selected Trees Sampled Along the Western Side of the Maxey Flats Restricted Area During July and August, 1983.

(See Figure 5 for Relative Tree Locations)

Tree No.	July Sampling	August Sampling
002	1.88 ± 04	8.60 ± 03
007	3.21 ± 05	3.02 ± 05
010	3.21 ± 04	2.43 ± 04
011	2.09 ± 04	1.40 ± 04
013	1.73 ± 04	7.50 ± 03
018	1.24 ± 04	1.40 ± 04
020	5.69 ± 04	2.41 ± 04

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